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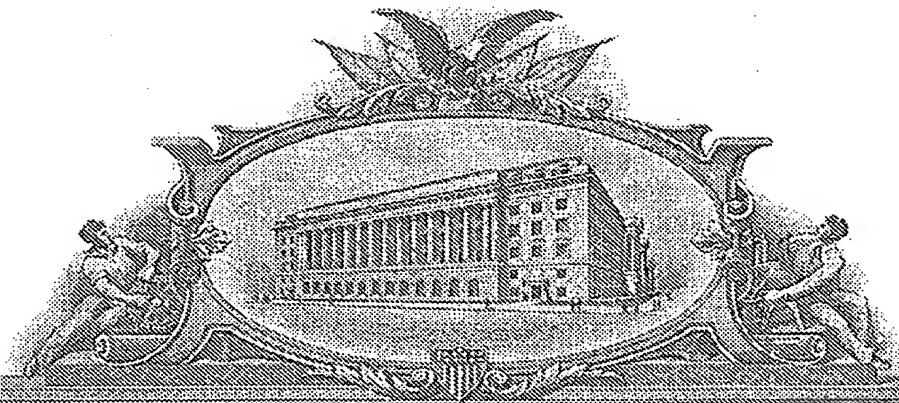
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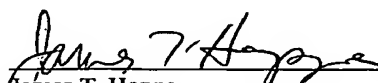
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PROVISIONAL APPLICATION COVER SHEET

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TITLE OF THE INVENTION (280 characters max)				
FUNCTIONALIZED ELASTOMERIC COMPOSITIONS				
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PROVISIONAL APPLICATION FILING ONLY

FUNCTIONALIZED ELASTOMER COMPOSITIONS

TECHNICAL FIELD

The invention relates to functionalized elastomer compositions comprised of olefin copolymers having chain end functionalized crystallizable or high T_g polyolefin sidechains grafted onto low crystallinity polyolefin backbones.

BACKGROUND AND SUMMARY OF THE INVENTION

Triblock and multi-block copolymers are well-known in the art relating to elastomeric polymers useful as thermoplastic elastomer ("TPE") compositions due to the presence of "soft" (elastomeric) blocks connecting "hard" (crystallizable or glassy) blocks. The hard blocks bind the polymer network together at typical use temperatures. However, when heated above the melt temperature or glass transition temperature of the hard block, the polymer flows readily exhibiting thermoplastic behavior. See, for example, G. Holden and N. R. Legge, *Thermoplastic Elastomers: A Comprehensive Review*, Oxford University Press (1987).

The best commercially known class of TPE polymers are the styrenic block copolymers (SBC), typically linear triblock polymers such as styrene-isoprene-styrene and styrene-butadiene-styrene, the latter of which when hydrogenated become essentially styrene-(ethylene-butene)-styrene block copolymers. Radial and star branched SBC copolymers are also well-known. These copolymers typically are prepared by sequential anionic polymerization or by chemical coupling of linear diblock copolymers. The glass transition temperature (T_g) of the typical SBC TPE is equal to or less than about 80-90°C, thus presenting a limitation on the utility of these copolymers under higher temperature use conditions. See, "Structures and Properties of Block Polymers and Multiphase Polymer Systems: An Overview of Present Status and Future Potential", S. L. Aggarwal, Sixth Biennial Manchester Polymer Symposium (UMIST Manchester, March 1976).

Insertion, or coordination, polymerization of olefins can provide economically more efficient means of providing copolymer products, both because of process

efficiencies and feedstock cost differences. Thus useful TPE polymers from olefinically unsaturated monomers, such as ethylene and $C_3 - C_8$ alpha-olefins, have been developed and are also well-known. Examples include the physical blends of thermoplastic olefins ("TPO") such as polypropylene with ethylene-propylene copolymers, and similar blends wherein the ethylene-propylene, or ethylene-propylene-diolefin phase is dynamically vulcanized so as to maintain well dispersed, discrete soft phase particles in a polypropylene matrix. See, N. R. Legge, "Thermoplastic elastomer categories: a comparison of physical properties", ELASTOMERICS, pages 14-20 (September, 1991), and references cited therein.

US4999403, herein incorporated in its entirety by reference, discloses graft copolymer compositions comprising a functionalized ethylene-alpha-olefin copolymer having polypropylene grafted thereto through one or more functional linkages. The disclosed process for preparing the graft copolymer compositions comprised combining functionalized ethylene-alpha-olefin copolymer with maleated polypropylene under conditions sufficient to permit grafting of at least a minor portion of the functionalized polymer with the polypropylene. It is well known in the art that the introduction of maleic acid functionality into a polymer through radical grafting results in a distribution of functionalities along the polymer backbone. The reaction of the resulting modified polypropylene with a functionalized elastomer will therefore result in irregular branching, potential for cross linking, and therefore inconsistent and/or undesirable properties.

It is desirable to prepare graft copolymer compositions with a controlled branching architecture, no cross linking, e.g. gel weight fraction less than 10%, preferably less than 5%, more preferably less than 3%, and most preferably less than 1%, measured in accordance with ASTM method D2765 and predictable and controllable properties.

We have now discovered olefinic compositions comprising a functionalized branched olefin copolymer containing sidechains which are also functionalized, said sidechains being derived from an olefin and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers, the copolymer

having A) a $T_g < -10^\circ\text{C}$ as measured by DSC; B) a $T_m > 100^\circ\text{C}$; C) an elongation at break of greater than or equal to 500%; D) a Tensile Strength of greater than or equal to 1,500 psi (10,300 kPa) at 25°C .; E) a TMA temperature $> 80^\circ\text{C}$, and F) an elastic recovery of greater than or equal to 50%.

As used herein, "functionalized olefin copolymers" refer to olefin polymers that have been modified to introduce elements other than carbon and hydrogen. Preferably at least about 30% of the polymer molecules have been modified. Suitable examples of "functionalized olefin copolymers" include maleic anhydride graft modified polyolefins (e.g. polyethylene or polypropylene), and amine terminated polyolefins.

Preferably, the functionalized sidechains in the olefinic composition have a T_g of less than -30°C and the T_m of the sidechains is greater than or equal to 100°C .

Also preferred are thermoplastic elastomer compositions wherein said functionalized branched olefin copolymer comprises functionalized sidechains derived from ethylene or propylene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers.

The functionalized branched olefin copolymer preferably can comprise functionalized sidechains derived from 4-methyl-1-pentene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers.

Another embodiment of the invention is a process for making a functionalized branched olefin copolymer comprising reacting a functionalized elastomer (such as a maleated elastomer) with an amine terminated olefin polymer. Preferably, the reacting step is performed in an extruder, more preferably the reacting step is performed in solution.

The functionalized branched olefin copolymer in the compositions can comprise a functionalized ethylene/alpha-olefin copolymer having a density of less than about 0.89 g/cc, wherein the functionality is capable of reacting with a primary amine, especially a

functionalized propylene/alpha-olefin copolymer having a density of less than about 0.87 g/cc, wherein the functionality is capable of reacting with a primary amine.

Preferably, the functionalized copolymer is formed from components comprising an unsaturated organic compound containing at least one olefinic unsaturation and at least one carboxyl group or at least one derivative of the carboxyl group selected from the group consisting of an ester, an anhydride and a salt. Preferably, the unsaturated organic compound is selected from the group consisting of maleic, acrylic, methacrylic, itaconic, crotonic, alpha-methyl crotonic and cinnamic acids, anhydrides, esters and their metal salts and fumaric acid and its ester and its metal salt. Maleic anhydride is most preferred.

In yet another embodiment of the invention, a thermoplastic elastomer composition derived from at least two functionalized olefin copolymers has been discovered, each copolymer derived from olefins capable of insertion polymerization and each copolymer having a T_m difference of at least 40°C, the composition having: A) a T_g < -10°C as measured by DSC; B) a T_m > 100°C; C) an elongation at break of greater than or equal to 500%; D) a Tensile Strength of greater than or equal to 1,500 psi (10,300 kPa) at 25°C.; E) a TMA temperature > 80°C, and F) an elastic recovery of greater than or equal to 50%, wherein at least one functionalized copolymer is chain end functionalized with a primary amine, especially wherein the two functionalized olefin copolymers are selected from the group consisting of maleated elastomer and amine terminated olefin polymers, further, wherein one of the functionalized olefin copolymers is selected from the group consisting of maleated elastomers, and one functionalized olefin copolymer is selected from amine terminated olefin polymers. Preferably, the composition has an additional T_g of greater than about 80°C.

In still another embodiment, a thermoplastic elastomer composition derived from at least two functionalized olefin copolymers is disclosed, each copolymer derived from olefins capable of insertion polymerization and each copolymer having a T_g difference of at least 40°C, more preferably at least 100°C, the composition having A) a T_g < -10°C as measured by DSC; B) an elongation at break of greater than or equal to 500%; C) a

Tensile Strength of greater than or equal to 1,500 psi (10,300 kPa) at 25°C.; D) a TMA temperature $> 80^{\circ}\text{C}$, and E) an elastic recovery of greater than or equal to 50%, wherein at least one functionalized copolymer is chain end functionalized with a primary amine, preferably wherein the two functionalized olefin copolymers are selected from the group consisting of maleated elastomer and amine terminated olefin polymers, further, wherein one of the functionalized olefin copolymers is selected from the group consisting of maleated elastomers, and one functionalized olefin copolymer is selected from amine terminated olefin polymers. Preferably, the composition has an additional T_g of greater than about 80°C .

In yet another embodiment, an olefin composition is disclosed which comprises a functionalized branched olefin copolymer containing functionalized sidechains derived from ethylene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers, the copolymer having A) at least one $T_g < -10^{\circ}\text{C}$ as measured by DSC, B) an elongation at break of greater than or equal to 500%; C) a Tensile Strength of greater than or equal to 1,500, psi (10,300 kPa) at 25°C .; D) a TMA temperature $> 80^{\circ}\text{C}$, and E) an elastic recovery of greater than or equal to 50%. Preferably, the composition has an additional T_g of greater than about 80°C .

DETAILED DESCRIPTION OF THE INVENTION

The thermoplastic elastomer compositions, and blends thereof, of this invention are comprised of branched copolymers wherein both the copolymer backbone and polymeric sidechains are derived from monoolefins polymerized under coordination or insertion conditions with activated transition metal organometallic catalyst compounds. The sidechains are copolymerized so as to exhibit crystalline, semi-crystalline, or glassy properties suitable for hard phase domains in accordance with the art understood meaning of those terms, and are grafted to a polymeric backbone that is less crystalline or glassy than the sidechains, preferably, substantially amorphous, so as to be suitable for the complementary soft phase domains characteristic of thermoplastic elastomer compositions.

The sidechains are comprised of chemical units capable of forming crystalline or glassy polymeric segments preferably under conditions of insertion polymerization. Known monomers meeting this criteria are ethylene, propylene, 4-methyl-1-pentene, and copolymers thereof, including ethylene copolymers with α -olefin, cyclic olefin or styrenic comonomers. Ethylene or propylene copolymer sidechains are preferable provided that the amount of comonomer is insufficient to disrupt the crystallinity. Suitable comonomers include $C_3 - C_{20}$ α -olefins or geminally disubstituted monomers, $C_5 - C_{25}$ cyclic olefins, styrenic olefins and lower carbon number ($C_3 - C_8$) alkyl-substituted analogs of the cyclic and styrenic olefins. Preferably, the sidechains can comprise from 90-100 mol % olefin (such as ethylene or propylene), and from 0-10 mol % comonomer, preferably 92-99 mol % olefin and 1-8 mol % comonomer, most preferably 95-98 mol % olefin and 2-5 mol % comonomer. The selection of comonomer can be based upon properties other than crystallinity disrupting capability, for instance, a longer olefin comonomer, such as 1-octene, may be preferred over a shorter olefin such as 1-butene for improved polyethylene film tear. For improved polyethylene film elasticity or barrier properties, a cyclic comonomer such as norbornene or alkyl-substituted norbornene may be preferred over an α -olefin.

The M_n of the sidechains are within the range of from greater than or equal to 1,500 and less than or equal to 75,000. Preferably the M_n of the sidechains is from 1,500 to 50,000, and more preferably the M_n is from 1,500 to 25,000. The number of sidechains is related to the M_n of the sidechains such that the total weight ratio of the weight of the sidechains to the total weight of the polymeric backbone segments between and outside the incorporated sidechains is less than 60%, preferably 10-40%, most preferably from 10-25%. Molecular weight here is determined by gel permeation chromatography (GPC) and differential refractive index (DRI) measurements. It will be readily understood by one of ordinary skill in the art, that physical properties of the sidechains are most conveniently determined from the material prior to reacting to become the actual sidechain. In this regard, the term "sidechain" may be thought of as equivalent to a sidechain precursor.

The molecular weight distributions of polyolefin, particularly ethylene, polymers are determined by gel permeation chromatography (GPC) on a Waters 150°C high temperature chromatographic unit equipped with a differential refractometer and three columns of mixed porosity. The columns are supplied by Polymer Laboratories and are commonly packed with pore sizes of 10^3 , 10^4 , 10^5 and 10^6 Å. The solvent is 1,2,4-trichlorobenzene, from which about 0.3 percent by weight solutions of the samples are prepared for injection. The flow rate is about 1.0 milliliters/minute, unit operating temperature is about 140°C and the injection size is about 100 microliters.

The molecular weight determination with respect to the polymer backbone is deduced by using narrow molecular weight distribution polystyrene standards (from Polymer Laboratories) in conjunction with their elution volumes. The equivalent polyethylene molecular weights are determined by using appropriate Mark-Houwink coefficients for polyethylene and polystyrene (as described by Williams and Ward in *Journal of Polymer Science, Polymer Letters*, Vol. 6, p. 621, 1968).

$$M_{\text{polyethylene}} = a * (M_{\text{polystyrene}})^b.$$

In this equation, $a = 0.4316$ and $b = 1.0$. Weight average molecular weight, M_w , is calculated in the usual manner according to the following formula: $M_j = (\sum w_i(M_{ij}))^j$. Where w_i is the weight fraction of the molecules with molecular weight M_i eluting from the GPC column in fraction i and $j = 1$ when calculating M_w and $j = -1$ when calculating M_n .

The backbone, or backbone polymeric segments, when taken together with an equivalent to the sidechain interruption, should have a lower T_m (or T_g if not exhibiting a T_m) than the sidechains. Thus it will preferably comprise segments of chemical units not having a measurable crystallinity, or having a T_g lower than -10°C . The backbone segments as taken together typically will have a T_m less than or equal to 80°C . and a T_g less than or equal to -10°C . Elastomeric backbones will be particularly suitable, such will typically be comprised of ethylene and one or more of C_3 - C_{12} alpha-olefins or diolefins, particularly propylene, 1-butene, and 1-octene. Other copolymerizable monomers include generally disubstituted olefins such as 4-methyl-1-pentene, hexene, isobutylene, cyclic

olefins such as cyclopentene, norbornene and alkyl-substituted norbornenes, and styrenic monomers such as styrene and alkyl substituted styrenes. Low crystallinity backbones are suitable, examples are high comonomer content ethylene copolymers (as described before), e.g., greater than 8 mol % comonomer.

As indicated above the mass of the backbone will typically comprise at least 40 wt % of the total polymer mass (i.e. that of the backbone and the sidechains together) so the backbone typically will have a weight-average molecular weight (M_w) of at least equal to or greater than about 50,000.

The branched olefin copolymers as a whole (i.e. comprising the above sidechains and backbones) will typically have an M_w equal to or greater than 50,000 as measured by GPC/DRI as defined for the examples. The M_w typically is less than 300,000, preferably less than 250,000.

The thermoplastic elastomer composition of the invention can be prepared by a process comprising reacting a functionalized (such as a maleated) elastomer with an amine terminated olefin polymer. The grafting process can be carried out in a homogeneous solution, a melt blend of the two component polymers, or in an extruder. The melt blending process is commonly performed using a twin-rotor mixer, preferably a twin-screw extruder having modular mixing sections, of sufficient length such as to achieve adequate mixing. Solution grafting, i.e. heating both components in a common solvent such as hydrocarbons, chlorinated and unchlorinated aromatics, at a temperature suitable to dissolve both materials and mixing until the desired grafting level is achieved. The polymer is recovered by removing the solvent. Preferably, a solvent is chosen such that the grafted copolymer precipitates from solution on cooling below 30 °C, and the polymer can be recovered by filtration. Suitable solvents include hydrocarbon mixtures such as Isopar®E sold by Exxon Chemical. Percentage of the polyolefin such as polypropylene which is grafted can vary from low levels such as 30% by weight of total polyolefin, but preferably is greater than 50%, most preferably greater than 65%, but can be as high as 100%. Grafting level can be determined by GPC methods.

Suitable maleation techniques include those described in USP 5,346,963 (Hughes et al.), USP 5,705,565 (Hughes et al.), USP 4,762,890 (Strait et al.), USP 4,927,888 (Strait et al.), USP 5,045,401 (Tabor et al.), and USP 5,066,542 (Tabor et al.), the disclosures of each of which is incorporated herein by reference.

Throughout the description above, and below, the phrase "chain-end" or "terminal" when referring to functionality means a functional group within 10 monomer units from the end of the polymer chain.

In one embodiment, propylene with chain end unsaturation, suitable as branches for a subsequent grafting reaction, can be prepared under solution polymerization conditions with metallocene catalysts suitable for preparing either of isotactic or syndiotactic polypropylene. These polymers may be converted to primary amine-terminated reagents by one of several methods. These methods include, inter alia, hydroformylation followed by conversion of the aldehyde or ketone to a primary amine and hydroformylation in the presence of a secondary amine followed by conversion of the resulting tertiary amine to a primary amine. Levels of amination can vary depending on desired product properties, but is typically greater than 50% (mole percent based on ¹H NMR of chain ends), more preferably greater than 70%, and can be as high as 100%.

Generally, for isotactic polypropylene, the stereorigid transition metal catalyst compound is selected from the group consisting of bridged bis(indenyl) zirconocenes or hafnocenes. In a preferred embodiment, the transition metal catalyst compound is a dimethylsilyl-bridged bis(indenyl) zirconocene or hafnocene. More preferably, the transition metal catalyst compound is selected from a series of pyridyl amine catalysts as disclosed in WO 2002/038628, USP 6,320,005 and USP 6,103,657, the disclosures of which are incorporated herein by reference.

The polypropylene sidechains of one embodiment of the present invention are preferably prepared in solution at a temperature from 110°C to 130°C. More preferably, a temperature from 110°C to 125°C is used. The pressures of the reaction generally can vary from atmospheric to 345 MPa, preferably to 182 MPa. The reactions

can be run batchwise or continuously. Conditions for suitable slurry-type reactions will also be suitable and are similar to solution conditions, except the reactions are typically carried out at lower temperatures. The polymerization is typically run in liquid propylene under pressures suitable to such.

Additionally the sidechains are prepared under suitable conditions such that greater than 50% of the chain end groups are unsaturated, preferably greater than 65%, most preferably greater than 80%, but can be as high as 100% (mole percent determined by ¹H NMR of end groups). Unsaturated end groups can include vinyl, vinylidene, vinylene, or mixtures thereof.

The thermoplastic elastomer compositions according to the invention will have use in a variety of applications wherein other thermoplastic elastomer compositions have found use. Such uses include, but are not limited to, those known for the styrene block copolymers, e.g., styrene-isoprene-styrene and styrene-butadiene-styrene copolymers, and their hydrogenated analogs. Such include a variety of uses such as backbone polymers in adhesive compositions and molded articles. These applications will benefit from the increased use temperature range, typically exceeding the 80-90°C limitation of the SBC copolymer compositions. The compositions of the invention will also be suitable as compatibilizer and impact modifier compounds for polyolefin blends. Additionally, due to the relatively high tensile strength, elasticity, and ease of melt processing, extruded film, coating and packaging compositions can be prepared comprising the invention thermoplastic elastomer compositions, optionally as modified with conventional additives and adjuvents. Further, in view of the preferred process of preparation using insertion polymerization of readily available olefins, the invention thermoplastic elastomer compositions can be prepared with low cost petrochemical feedstock under low energy input conditions (as compared to either of low temperature anionic polymerization or multistep melt processing conditions where vulcanization is needed to achieve discrete thermoplastic elastomer morphologies).

EXAMPLES

The following examples are given to illustrate various embodiments of the invention. They do not intend to limit the invention as otherwise described and claimed herein. All numerical values are approximate. In the following examples, various polymers were characterized by a number of methods. Performance data of these polymers were also obtained. Most of the methods or tests were performed in accordance with an ASTM standard, if applicable, or known procedures.

Isopar®E hydrocarbon mixture is obtained from Exxon Chemicals. Rac-[Dimethylsilane-diylbis(1-(2-methyl-4-phenyl)indenyl)]zirconium (trans,trans-1,4-Diphenyl-1,3-butadiene) is prepared according to USP 6,465,384, the disclosure of which is incorporated herein by reference, especially example 15. Bis(hydrogenated-tallowalkyl)methylammonium tetrakis(pentafluorophenyl)borate is prepared according to USP 5,919,983, the disclosure of which is incorporated herein by reference, especially example 2. PMAO-IP is obtained as a toluene solution from Akzo Chemicals and is used without further purification.

Unless indicated otherwise, the following testing procedures are to be employed:

A. Tensile Testing At Room Conditions. Tensile testing is done using ASTM D-1708 with microtensile bars cut to the sample specifications. The cross-head speed is set to 127 mm/min. (5 inches/min.). Testing environment is not to ASTM standards for temperature and humidity. Samples were tested as is and were not conditioned according to ASTM D-1708.

B. Procedure for Tensile Hysteresis Tensile hysteresis is measured using the geometry outlined in ASTM D1708. The gauge length is 22.25 mm long by 4.8 mm wide. The loading and unloading strain rate is 500%/mm. The test procedure is carried out as follows: The sample is loaded with Mylar in grips and the load is zeroed. The sample is then pulled to 100% strain The sample is retracted to 0% strain and reloaded to positive

load Permanent set is the strain at which the load becomes zero upon reloading. The elastic recovery is defined as 100% minus the permanent set.

C. DSC Differential Scanning Calorimetry (DSC) measurements are performed on a TA Instruments Q1000. Heat the sample in DSC to 30°C (at approximately 100°C/min) above the melting point. Keep isothermal for 3 minutes to ensure complete melting. Cool the sample at 10°C/min to -40°C. Keep the sample isothermal for three minutes to stabilize. Melting (from second heat) and crystallization temperatures are recorded from the peak temperatures of the endotherm and exotherm, respectively. Glass transition temperature is taken as the temperature at the inflection point of the change in heat capacity.

D. TMA A Perkin Elmer TMA 7 (Thermomechanical Analyzer) is loaded with samples with a thickness of 2 to 4 mm. A flat-headed needle with a load of one Newton was placed against the sample at room temperature. The temperature is ramped at 5°C/min from 25°C to 190°C. The test is stopped before 190°C if the needle has penetrated 1 mm into the sample. The TMA temperature is defined as the temperature at which the sample penetration reaches 1 mm.

Polypropylene synthesis. A stirred, one gallon (3.79 L) autoclave reactor is charged with 1400g Isopar®E hydrocarbon solvent and 580g propylene. The reactor is heated to the desired temperature (110°C – 125°C). The catalyst system is prepared in a drybox by combining together rac-[Dimethylsilane-diylbis(1-(2-methyl-4-phenyl)indenyl)]zirconium (trans,trans-1,4-Diphenyl-1,3-butadiene), bis(hydrogenated-tallowalkyl)methylammonium tetrakis(pentafluorophenyl)borate, and AKZO PMAO-IP in a 1:1.1:38 molar ratio, with additional solvent to give a total volume of 17 ml. The activated catalyst is injected into the reactor. The reactor temperature is maintained constant by cooling the reactor as required. After 10 minutes the hot solution is transferred into a nitrogen purged resin kettle. An additive solution containing a phosphorus stabilizer and phenolic antioxidant (Irgaphos 168 and Irganox 1010 in toluene in a 2:1 weight ratio) is added to provide a total additive concentration of about

0.1 wt percent in the polymer. The polymer is dried in at vacuum oven at 70°C over night.

Example 1 Preparation of amine terminated polypropylene

A] Hydroformylation of Olefin-Terminated Polypropylene. A one-gallon Parr reactor is charged with olefin-terminated polypropylene prepared above (244g), and toluene (1472 g, 1702 mL). The reactor is purged with 1:1 syn gas and then vented. Via cannula transfer, 128g of a catalyst solution is charged. The catalyst solution consisted of dry, deoxygenated THF (165g, 186 mL), Rh(CO)₂(acac) (2.47g, 9.57 mmol), and tris(2,4-di-*t*-butylphenyl)phosphite (30.12g, 46.6 mmol) (L/Rh = 4.87; 4997 ppm Rh). The reactor is pressurized to 200 psi with 1:1 syn gas and heated to 80°C, then pressurized to 300 psi and heated to 100°C. After 4 hours, the reactor is vented, dumped hot and washed with hot toluene. The polymer is precipitated by pouring into methanol, and then washed with additional methanol, and dried in vacuo. (NOTE: It is later determined that washing with acetone leads to more complete removal of catalyst residues). 232g (95%) of white powder were recovered. ¹H NMR resonances between δ 9.6 – 9.9 were assigned to aldehyde hydrogens.

B] In a nitrogen atmosphere, a three-liter flask is charged with tetrahydrofuran ("THF") (1000 mL), formyl-terminated polypropylene (200g), and triethylamine (4.65 mL, 33.3 mmol). A solution of , hydroxylammonium chloride (1.72g, 26.7 mmol) in 200 mL THF was placed in an addition funnel attached to the three-liter flask. The hydroxylamine hydrochloride solution is added dropwise over ~1 hour to the stirring polymer slurry. At this time, the reaction mixture is stirred and heated to 60°C for six hours. After cooling to room temperature, the polymer is washed sequentially with water, methanol, and acetone. ¹H NMR resonances between δ 6.3 – 6.8 were assigned to oxime hydrogens.

C] Reaction of Oxime-Terminated Polypropylene to Form Amine-Terminated Polypropylene. In a nitrogen filled glove box, a 2-L flask is charged with 100g of the oxime-terminated polypropylene prepared above and 800 mL dry THF. To the slurry is added 60 mL of a 1M solution of LiAlH₄ in THF. The solution is heated to reflux for 4 hours. The solids dissolved on heating to form a homogeneous solution, and over the

course of the reaction a grey precipitate formed. The polymer is allowed to cool to a gel and is brought out of the box. The polymer/solvent gel is added to 1L of MeOH with stirring. Some gas evolution is observed as residual LiAlH_4 is consumed. The polymer is stirred for 30 minutes, collected on a fritted funnel, washed twice with 500 mL MeOH, and aspirated to a free flowing powder. The powder is dried in a vacuum oven at 50°C over night.

Example 2 Preparation of the functionalized branched olefin copolymer

Samples of Ethylene-Octene Copolymer grafted with Maleic Anhydride (DuPont Fusabond NMN-4940 is a 0.87 g/cc density EO-g-MAH 1 MI with about 1 wt% MAH) were mixed with amine-terminated polypropylene in a Haake Rheocord 9000 mixer. A total of 140 grams of EO-g-MAH is melted at 170°C in a Haake R3000 bowl with a sample volume of 310 ml at 30 RPM. A total of 60 grams of amine-terminated PP is slowly added and each aliquot is allowed to react to completion. The reaction is monitored via an increase in torque. Once all of the PP is added, the graft copolymer is melt mixed for another five minutes.

Properties

Sample	Tensile Strength, psi	Elongation @ Break, %	Elastic Recovery, %	TMA, °C
Blend*	1220	720	69	79
Graft Copolymer**	2490	800	78	110

*Blend: physical blend of EO-g-MAH and iPP ** Graft Copolymer: example of this invention; graft copolymer of EO-g-MAH iPP and NH_2 -t-iPP

We claim:

1. An olefinic composition comprising a functionalized branched olefin copolymer containing sidechains which are also functionalized, said sidechain being derived from an olefin and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers, the copolymer having A) a $T_g < -10^\circ\text{C}$ as measured by DSC; B) a $T_m > 100^\circ\text{C}$; C) an elongation at break of greater than or equal to 500%; D) a Tensile Strength of greater than or equal to 1,500 psi (10,300 kPa) at 25°C ; E) a TMA temperature $> 80^\circ\text{C}$, and F) an elastic recovery of greater than or equal to 50%.
2. The composition of Claim 1 wherein the sidechain is derived from ethylene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers.
3. The olefinic composition of claim 1 where the T_g of the functionalized sidechains is less than -30°C , and the T_m of the sidechains is greater than or equal to 100°C .
4. The composition of claim 1 wherein said functionalized branched olefin copolymer comprises functionalized sidechains derived from propylene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers.
5. The composition of claim 1 wherein said functionalized branched olefin copolymer comprises functionalized sidechains derived from 4-methyl-1-pentene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers.
6. A process of making a functionalized branched olefin copolymer comprising reacting a maleated elastomer with an amine terminated olefin polymer.
7. The process of claim 6, wherein the reacting step is performed in an extruder.
8. The process of claim 6, wherein the reacting step is performed in solution.

9. The composition of claim 1 wherein said functionalized branched olefin copolymer comprises a functionalized ethylene/alpha-olefin copolymer having a density of less than about 0.89 g/cc, wherein the functionality is capable of reacting with a primary amine.

10. The composition of claim 1 wherein said functionalized branched olefin copolymer comprises a functionalized propylene/alpha-olefin copolymer having a density of less than about 0.87 g/cc, wherein the functionality is capable of reacting with a primary amine.

11. The composition of claim 1 wherein the functionalized copolymer is formed from components comprising an unsaturated organic compound containing at least one olefinic unsaturation and at least one carboxyl group or at least one derivative of the carboxyl group selected from the group consisting of an ester, an anhydride and a salt.

12. The composition of claim 11 wherein the unsaturated organic compound is selected from the group consisting of maleic, acrylic, methacrylic, itaconic, crotonic, alpha-methyl crotonic and cinnamic acids, anhydrides, esters and their metal salts and fumaric acid and its ester and its metal salt.

13. A thermoplastic elastomer composition derived from at least two functionalized olefin copolymers, each copolymer derived from olefins capable of insertion polymerization and each copolymer having a T_m difference of at least 40°C, the composition having A) a $T_g < -10^\circ\text{C}$ as measured by DSC; B) a $T_m > 100^\circ\text{C}$; C) an elongation at break of greater than or equal to 500%; D) a Tensile Strength of greater than or equal to 1,500 psi (10,300 kPa) at 25°C; E) a TMA temperature $> 80^\circ\text{C}$, and F) an elastic recovery of greater than or equal to 50%, wherein at least one functionalized copolymer is chain end functionalized with a primary amine.

14. A thermoplastic elastomer composition derived from at least two functionalized olefin copolymers, each copolymer derived from olefins capable of insertion polymerization and each copolymer having a T_g difference of at least 100°C, the composition having A) at least one $T_g < -10^\circ\text{C}$ as measured by DSC; B) an elongation at break of greater than or equal to 500%; C) a Tensile Strength of greater than or equal to

1,500 psi (10,300 kPa) at 25°C; D) a TMA temperature > 80°C, and E) an elastic recovery of greater than or equal to 50%, wherein at least one functionalized copolymer is chain end functionalized with a primary amine.

15. The composition of claims 13 or 14, wherein the composition has an additional T_g of greater than about 80°C.

16. The composition of claims 13 or 14, wherein the two functionalized olefin copolymers are selected from the group consisting of maleated elastomer and amine terminated olefin polymers.

17. The composition of claims 13 or 14, wherein one of the functionalized olefin copolymers is selected from the group consisting of maleated elastomers, and one functionalized olefin copolymer is selected from amine terminated olefin polymers.

18. An olefin composition comprising a functionalized branched olefin copolymer containing functionalized sidechains derived from ethylene and at least one chain end primary amine functional group, optionally with one or more copolymerizable monomers, the copolymer having A) at least one T_g < -10°C as measured by DSC, B) an elongation at break of greater than or equal to 500%; C) a Tensile Strength of greater than or equal to 1,500, psi (10,300 kPa) at 25°C.; D) a TMA temperature >80°C, and E) an elastic recovery of greater than or equal to 50%.

19. The composition of claim 18, wherein the copolymer further comprises an additional T_g of greater than about 80°C.

From the INTERNATIONAL BUREAU

PCTNOTIFICATION CONCERNING
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(PCT Administrative Instructions, Section 411)

To:

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Date of mailing (day/month/year) 17 April 2005 (17.04.2005)	
Applicant's or agent's file reference 63610B	IMPORTANT NOTIFICATION
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Applicant	DOW GLOBAL TECHNOLOGIES INC. et al

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<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
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